Heterogeneous Nucleation and Hot Spots at Grain Boundaries in a RDX Bicrystal

Marc J. Cawkwell, Thomas D. Sewell, T-14; Kyle J. Ramos, Daniel E. Hooks, DE-9

A grain boundary is an interface between two crystals with different orientations. Grain boundaries are structural and elastic heterogeneities in a polycrystal and strongly influence a number of material properties [1,2]. Using non-equilibrium molecular dynamics (NEMD) simulations, we have studied the effects of a grain boundary on the response to shock loading of the energetic molecular crystal cyclotrimethylene trinitramine (RDX).

We constructed a bicrystal, with grains oriented as shown in Fig. 1, that measured 39.7 x 5.4 x 100.0 nm. Two parallel grain boundaries were created upon the application of periodic boundary conditions. The system was thermalized to 300 K and zero external pressure. The resulting grain boundaries displayed a period of about 13.8 nm over which the structure transitioned from semi-coherent to disordered. The bicrystal was impacted onto a stationary piston at a particle velocity of 500 m/s, generating a shock wave that propagated parallel to the plane of the grain boundaries.

Earlier NEMD studies of the propagation of shock waves parallel to [010] had revealed a shock-induced phase transformation [3]. These simulations employed a crystal free from pre-existing defects, hence the new phase nucleated homogeneously. Shock pressures around 10 GPa were required to nucleate even small amounts of the new phase. However, recent flyer-plate-driven shock wave experiments suggest that the phase transformation occurs at pressures of approximately 4 GPa. During our NEMD simulations of the bicrystal, we found that the new phase nucleates heterogeneously within the [010]-oriented grain from the disordered sections of the grain boundaries at a shock pressure around 4.5 GPa (Fig. 2). Hence, we attribute the discrepancy between our earlier theoretical predictions and recent experiments to the existence of heterogeneities that are inevitably present in bulk RDX single crystals.

We found no evidence for grain boundary sliding arising from the different shock propagation velocities in the two grains. However, we did measure a significant increase in intramolecular temperatures at the grain boundaries. This is shown in Fig. 3 where we plot intramolecular temperatures calculated in slices parallel to the plane of the grain boundaries at the time of maximum compression during shock loading. The grain boundaries are 50-60 K hotter than the bulk crystals. Hence, grain or particle boundaries in energetic materials are hot spots that increase detonation sensitivity even in the absence of frictional heating due to interfacial sliding.

For further information contact Marc J. Cawkwell at cawkwell@lanl.gov.

[1] A.P. Sutton, R.W. Balluffi, *Interfaces in Crystalline Materials,* Oxford University Press, Oxford (1995).
[2] I. Alber et al., *Phil. Trans. R. Soc. Lond. A*, **339,** 555 (1992).
[3] M.J. Cawkwell, T.D. Sewell, *"New Shock-Induced Phase Transformation in RDX,"* Los Alamos National Laboratory report LALP-07-041 (2007).

Funding Acknowledgments
- Joint memorandum of understanding (MOU) between the Department of Energy and the Department of Defense
- Department of Energy, National Nuclear Security Administration
- Los Alamos National Laboratory Directed Research and Development Program
- Office of Naval Research

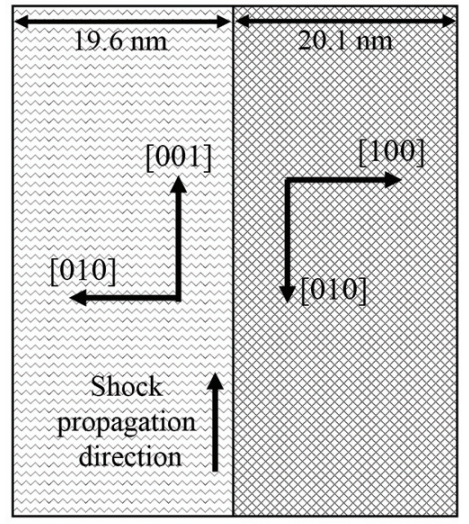

Fig. 1. Schematic illustration of the bicrystal employed in the NEMD simulation.

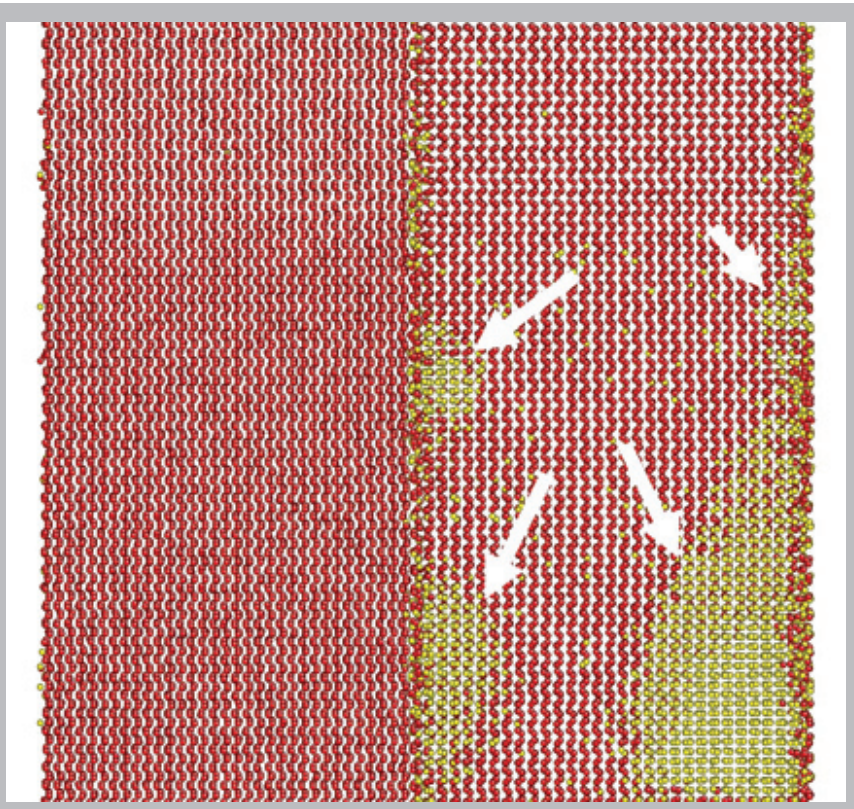

Fig. 2. Section from the simulation cell during shock loading at a particle velocity of 500 m/s. The centers of mass of molecules identified as being in the α-RDX environment are colored red while molecules belonging to the shock-induced phase are colored yellow. Arrows identify four heterogeneous nucleation events.

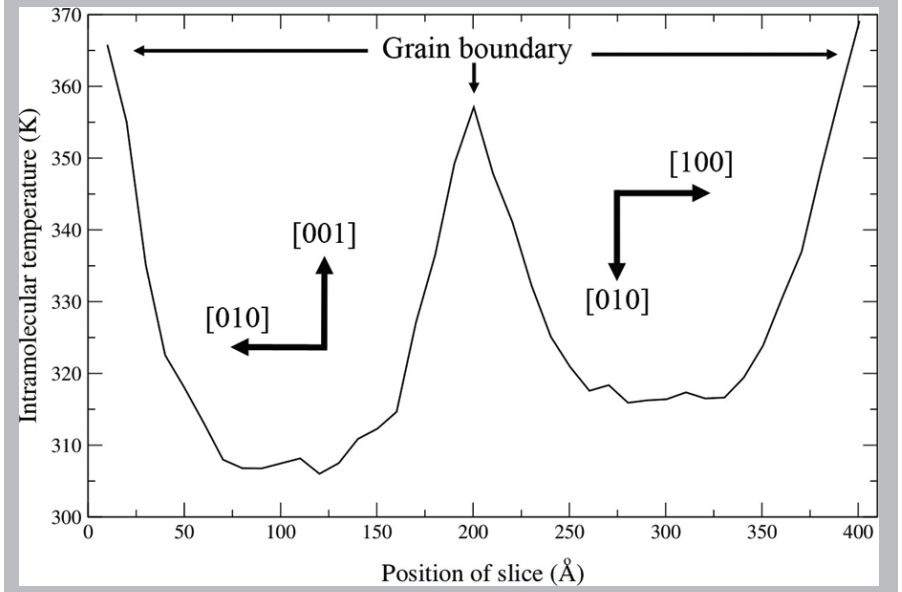

Fig. 3. Average intramolecular temperature calculated within 10-Å thick slices parallel to the grain boundary plane. The simulation cell was at the time of maximum compression in the NEMD simulation.